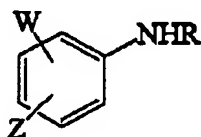


WHAT IS CLAIMED IS:

1. (Currently amended) A process for the direct synthesis of mono-N-substituted anilines having the general formula (I)



(I)

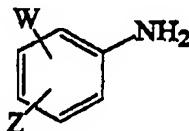
5 wherein

R indicates a linear or branched saturated carbon chain, preferably comprising 1 to 7 carbon atoms, an unsaturated carbon chain with the double carbon-carbon link also in the allyl position (2,3) with respect to the nitrogen atom of the amines (I), comprising 3 to 7 carbon atoms, or a benzyl group or a benzyl group substituted at the aromatic ring with methyl and ethyl radicals;

W is selected from the group consisting of [-H], -OH, -CH₂OH, -COOH and -CONH₂ and can be ortho, meta or para with respect to the carbon atom to which the nitrogen atom is attached;

15 Z is selected from the group consisting of -H, -halogen, -alkyl, -alkoxy, -NO₂ and -CN; ~~provided that W and Z are not simultaneously H,~~

said process comprising the step of reacting, in the presence of a solvent, a compound having the general formula (II):

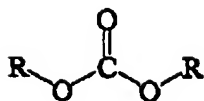


(II)

20 wherein W and Z are as defined above,

with an organic carbonate selected from the group consisting of compounds having the general formula (III)

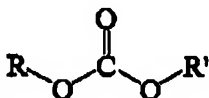
15



(III)

wherein R is as defined above,
and compounds of general formula (IV)

(IV)



5 wherein R' is selected from the group consisting of $\text{CH}_3(\text{OCH}_2\text{CH}_2)_n$ -
with $n \geq 2$ and branched or linear alkyl radicals that have at least three
carbon atoms;

in the presence of a faujasite selected from the group comprising X-
faujasite exchanged with alkaline metals and Y- faujasite exchanged with
10 alkaline metals.

2. (Previously submitted) The process according to claim 1, wherein
the organic carbonate has the general formula (III).

3. (Previously submitted) The process according to claim 1, wherein
the organic carbonate has the general formula (IV).

15 4. (Previously submitted) The process according to claim 1, wherein
the radical R is selected from the group consisting of methyl, ethyl, allyl and
benzyl.

5. (Previously submitted) The process according to claim 1, wherein
said faujasite is present in a ratio between 1:10 and 3:1 with respect to the
20 compound having the formula (I).

6. (Previously submitted) The process according to claim 5, wherein
said faujasite is present in a ratio between 1:1.5 and 1:1 with respect to the
compound having the formula (I).

7. (Previously submitted) The process according to claim 1, wherein
25 said faujasite is Y-faujasite exchanged with sodium.

8. (Previously submitted) The process according to claim 1, wherein

reaction conditions, particularly as regards the temperature, are particularly mild, and this aspect is a considerable advantage.

The achievement of high yields and excellent chemoselectivity even in these mild conditions has been achieved here mainly by way of the combined use of the carbonates cited above and of faujasites.

This last class of compounds indicates solids of the aluminosilicate type, which due to their structure can contain various types of ions of alkaline metals, alkaline-earth metals and/or transition metals.

Preferably, according to the present invention X-faujasites and Y-faujasites, both exchanged with alkaline metals, and even more preferably Y-faujasites exchanged with sodium, are used as catalysts.

As regards the quantities in which they are used, it has been found that it is possible to vary the percentages considerably while keeping, however, an interval of weight ratio: Y-faujasite / aniline substrate between 1:10 and 3:1 and more preferably between 1:1.5 and 1:1.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Further characteristics and advantages of the process according to the present invention will become better apparent from the following detailed description of some preferred but not exclusive embodiments thereof.

Examples.

In the performed experimental tests reported here, the quantity of organic substrate (functionalized aniline) was varied over a range comprised between 0.5 g and 10 g, preferably 1g.

Examples 1 to 7 and 12 refer to reactions performed in conventional reaction flasks and at atmospheric pressure, while examples 8 to 11 relate to processes performed in an autoclave.

Example 1

N-methyl-p-aminophenol.

The p-aminophenol (1 g) is dissolved in a mixture consisting of dimethyl carbonate (hereafter referenced as DMC) (10 ml) and DME (35

ml).

The NaY-faujasite (1 g) is added to the resulting mixture and heating to 90°C is performed.

The reaction, which is monitored by gas chromatography optionally
5 associated with mass spectrometry, is completed after 7 hours, providing a practically quantitative conversion.

At this point the solid catalyst is eliminated by filtration and the monomethylated product can thus be recovered.

Instrumental analysis performed after solvent evaporation yielded a
10 selectivity value of 99% for the N-monomethylated product and a total yield of 91%.

Example 2

N-methyl-o-aminophenol

The o-aminophenol (1 g) is dissolved in pure DMC (30 ml).

15 In this case, no co-solvent is added.

The NaY-faujasite (1 g) is added to the resulting mixture and heating to 90°C is performed.

The reaction, which is monitored by means of gas chromatography optionally associated with mass spectrometry, is completed after 3 hours,
20 providing a practically quantitative conversion.

At this point the solid catalyst is eliminated by filtration and the monomethylated product can thus be recovered.

Instrumental analysis performed after solvent evaporation reveals a total yield of 99% of N-monomethylated product.

25 Example 2a

The method of example 2 is repeated, but in this case the quantity of NaY-faujasite is limited to 1/10 of the quantity of o-aminophenol (again 1 g).

The reaction is completed after 38 hours, giving a total yield of 93% of
30 N-monomethylated product.